VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the Specification:

Please replace the paragraph beginning on page 4, line 17 with the following:

The present inventors have chosen an enhancement of a known technique for achieving calibration-free mass spectrometry measurements. The known technique is Isotope Dilution Mass Spectrometry, hereinafter IDMS. In IDMS, one takes advantage of well-known, naturally-occurring isotope ratios in essentially all elements. For example, it is well-known that the average atomic mass of copper (Cu) in the natural state is 63.546, consisting of isotopes 62.9396 at 69.2% and 64.9276 at 30.8%. Very generally speaking, assuming an analysis for Cu content is required, one takes a sample of the solution to be analyzed, spikes the sample with an enriched standard solution having a substantially different isotope ratio than the naturally-occurring ratio, ~~introduces~~introducing the spiked sample to a mass spectrometer, and records the measured mass ratio between the isotopes. The measurement is going to differ from the naturally-occurring ratio and the standard spike ratio, and from the measured ratio, knowing the quantity of the original sample, and the quantity of the spike solution, one can calculate the single unknown, that being the concentration of Cu in the original sample.

On page 18, line 8 insert the following paragraphs:

-- It is an object of the present invention to provide an analytical apparatus and an associated method to monitor fluid systems by extracting raw samples and modifying the same chemically and/or as to concentration with subsequent analytical analysis being performed with the assistance of automated computerized control.

It is a further object of the present invention to provide such a system that employs isotopic ratios in effecting such determinations on liquid samples.

It is another object of the present invention to provide such a system that has unique liquid handling subsystems, which provide for automated continuous processing.

It is another object of the present invention to provide such systems that facilitate decision control over raw sample amounts, as well as chemical modifications and dilutions thereto.

It is another object of the present invention to provide such a system that is adapted for online or adjacent analysis with respect to industrial process wet-baths.

These and other objects of the invention would be more fully understood

from the following detailed description of the invention on reference to the illustrations appended hereto--.

Please replace the paragraph beginning on page 18, line 31 with the following:

Speciated Isotope Dilution Mass Spectrometry (SIDMS), as described in ~~the '259 patent to Kingston~~Kingston United States Patent 5,414,259 referenced above, has been developed to assess the quantification of species and also their transformations. In SIDMS a predetermined species is specifically isotopically labeled and introduced to accomplish such measurements. The species of interest is previously known and specifically evaluated. In this invention the labeled species (frequently multiple species simultaneously) are being created in solution and are not previously determined in composition and structure until evaluated for structural information. Quantification is of the elemental ion and speciation information first established in-process through dynamic equilibrium established with a non-complexing salt of an enriched stable isotope in real time and in-process.

Please replace the paragraph beginning on page 24, line 29 with the following:

More specifically extraction modules 301, 305, 309, 313, and 317 are shown paired with wet-baths 303, 307, 311, 315, and 319, respectively. Each extraction module is connected to its wet bath by at least one fluid conduit, as module 301 is shown connected to bath 303 by exemplary conduit 321.

Please replace the paragraph on page 27, line 5 with the following:

There are three chemical introduction units labeled as units 1, 2, and 3, and labeled also element numbers 331, 333 and 335 respectively. These modules are for introducing chemicals for any of a range of purposes as ~~desceried~~described briefly above, such as to neutralize strong acid or base components of introduced samples. Each of these units is charged with appropriate chemicals for the kinds of baths to which the system will be devoted. Further, each is capable of introducing precisely measured quantities of these chemicals, under control of the computerized control system described briefly above. These chemical constituents are provided on demand to switching valve SV2, and thence to mixer 1 (341) via conduit 365.

Please replace the paragraph beginning on page 30, line 6 with the following:

In operation, plunger ~~434~~515 is withdrawn with valve 511 connecting the syringe to path 509 through valve 507 switched to provide the proper path and open valve 503. A precise volume of the chemical solution is drawn into the syringe, and may then be injected at the appropriate time and at an appropriate rate by switching valve 511 to provide

a path between the syringe and conduit 337 (see Fig. 3), which conducts the chemical introduced to SV2 and then mixer 1. A source of UPW (ultra pure water) is also made available through three-way valve 507, and nitrogen through valve 505. During syringe loading valve 505 is opened to prevent a vacuum being drawn on the reservoir by the syringe action.

Please replace the paragraph beginning on page 31, line 22 with the following:

Communication between primary syringe 601 and the arrangement of secondary syringes and other elements is through a switching valve 609. In the event 1ppm spike is needed, for example, syringe 619 is used through ~~valve~~valves 621 and 623 to draw a precise amount from reservoir 611, and the 1ppm volume is then transferred to syringe 601 by switching valve 621 to the right-hand position, and switching valve 609 to transfer the volume through valve 603 and into syringe 601. When the spike is needed for a sample to be analyzed, switching valve 609 is positioned with valve 603 open to syringe 601 to allow syringe 601, by driving its plunger, to move the spike into conduit 355 and on to SV3 (see Fig. 3).

Please replace the paragraph beginning on page 32, line 8 with the following:

Assume that a 33 ppb spike is required. The first step is the same as for the 1 ppm spike as described above, that is, 1 ppm spike is drawn from reservoir 611 into syringe 619. During this operation valve 613 may be opened to a controlled-pressure nitrogen source for the same purposes as previously described. While the 1 ppm solution is drawn into syringe 619, valves 641, 639, and 667 are set to connect syringe 645 to reservoir 669, and a precise volume of dilution solution is drawn into syringe 645. Now valve 667 is closed, and valves 641, 639, 637, and 635 are reset to straight-through, connecting syringe 645 to mixer 631. At the same time, after syringe 619 is filled with the 1 ppm solution, valves 621 and 623 ~~re-set~~reset to connect syringe 619 to mixer 631. Syringes 619 and 645 are now operated in concert with syringe 643 with valves 625 and 627 set straight-through, and the volume of 1 ppm solution from syringe 619 is mixed with the dilution solution in syringe ~~645through~~645 through mixer 631 and into syringe 643. At the end of this operation accomplished through valve manipulation and manipulation of the appropriate syringe plungers, syringe 643 has a precise spike at 33 ppb.

Please replace the paragraph beginning on page 33, line 12 with the following:

Fig. 7 is a block diagram of a control system for an instrument in an embodiment of the present invention. Instrument 707 in Fig. 7 represents an instrument as

shown abstractly in Fig. 2. With the exception of a computer station 215, comprising a computer 705 and a display unit 703, all of the components of the instrument are housed in a common cabinet, which may be positioned in a convenient location to fluid systems to be monitored, such as in a maintenance area outside the strict clean-room environment in a semiconductor fab. Computer 215 is analogous to the element with the same number in Fig. 2, and can be such as a high-end PC. Computer 215 is connected by a communication link 704 to the instrument 707, and this link can be any of several types, such as a serial link or a parallel communication link. Other links are also possible.

Please replace the paragraph beginning on page 34, line 1 with the following:

Microprocessor 217 is connected to an I/O interface 709 for controlling the fluid-handling and other elements of the instrument. It is well-known in the art that drive motors as used for such as syringe plungers, solenoids to drive diaphragms for pinch valves, multi-path switching valves, and the like, are all driven by AC or DC signals providing the voltage and current necessary to drive the specific elements. The I/O interface accepts computer-level signals from microprocessor 217 for manipulating elements of the instrument, and provides the correct signals as needed for each element in the voltage, current, and time required. There are also in many instances signals delivered back to I/O interface 709 from sensors in the instrument, such as position sensors on syringe plungers, which are translated to appropriate computer voltage and provided to microcontroller 217.

In the Claims:

1. (Amended) An analytical apparatus to monitor fluid systems, comprising:

at least one extraction module having a ~~raw-sample~~ raw sample reservoir connected by input fluid conduit to individual ones of said fluid systems, to extract raw samples for analysis from said fluid systems;

one or more modification modules comprising additive materials to modify the extracted raw samples prior to analysis;

an analytical device to receive at least a portion of said raw samples in an ordered sequence, and to determine concentration of at least one constituent of said sample portion;

fluid-handling apparatus for transferring fluid through the analytical apparatus; and

a computerized control and management system to manage operations of component modules and devices, and to report analytical results;

characterized in that the control and management system coordinates extraction of

raw samples, modification as ~~needed~~desired, introduction of fluids to the analytical device, and reporting of analytical results ~~on a continuous basis over plural cycles~~.

4. (Amended) The analytical apparatus of claim 3 wherein the processing environment is a fabrication system ~~(fab)~~ devoted to semiconductor integrated circuit ~~(IC)~~ manufacturing.

5. (Amended) The analytical apparatus of claim 3 wherein the modification modules comprise chemicals for altering the pH of extracted liquids.

10. (Amended) The analytical apparatus of claim 3 wherein the ~~raw-sample~~ raw sample reservoir in the at least one extraction module is connected to a vacuum apparatus through a remotely-operable valve, to draw a relative vacuum in the reservoir to draw material from the fluid system connected by said input fluid conduit to the reservoir.

11. (Amended) The analytical apparatus of claim 10 wherein the ~~raw-sample~~ raw sample reservoir is generally vertically-oriented and the conduit from the wet-bath enters at a height on the reservoir such that a liquid sample drawn into the reservoir may have a gas gap over the sample.

~~16~~ 106. (Amended) The analytical apparatus of claim 15 wherein the precision translation mechanism comprises a ~~precision~~ stepper motor.

~~22~~ 107. (Amended) The analytical apparatus of claim 21 wherein the control and management system is ~~enabled~~structured to vary the rate of plunger translation.

23. (Amended) The analytical apparatus of claim 20 wherein the syringe is connected through a three-way switching valve to the intermediate conduit to the chemical reservoir and to an output conduit to provide the precise volume sample to other elements of the analytical apparatus.

24. (Amended) The analytical apparatus of claim 20 wherein the portions of the solution provided to other elements are provided through a mixer having two or more inputs to one output, with one input connected to the chemical ~~alteration~~modification module and another

connected to another module in the analytical apparatus.

28. <u>(Amended)</u> The analytical apparatus of claim 27 wherein the syringe comprises a plunger driven by the control and management system through a ~~precision~~ translation mechanism, enabling precision volume control.

~~29~~<u>108</u>. <u>(Amended)</u> The analytical apparatus of claim 28 wherein the control and management system is ~~enabled~~<u>structured</u> to vary <u>the</u> rate of plunger translation.

29. <u>(Amended)</u> The analytical apparatus of claim 28 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor.

31. <u>(Amended)</u> The analytical apparatus of claim 26 wherein the portions provided to other elements are provided through a mixer having two or more inputs to one output, with one input connected to the chemical ~~alteration~~<u>modification</u> module and another connected to another module in the analytical apparatus.

33. <u>(Amended)</u> The analytical apparatus of claim 27 wherein the isotope reservoir contains an isotope mixture in a solvent at a specific concentration, and further comprising a solvent reservoir in addition to the at least one isotope reservoir, the solvent reservoir containing the solvent common to the isotope mixture in the isotope reservoir, and a system of syringes, connecting conduits and mixers, enabling portions of the isotope mixture to be diluted ~~to lesser concentrations~~ before being provided to other elements of the analytical apparatus.

34. <u>(Amended)</u> The analytical apparatus of claim 33 wherein the isotope reservoir holds an isotope mixture in a concentration ~~if~~<u>of about</u> 1 part ~~isotope~~<u>isotopes</u> in one million parts of the mixture ~~(1ppm)~~, and the system of syringes, connecting conduits and mixtures enables dilution of six orders of magnitude to <u>about</u> one part per trillion ~~(1ppt)~~.

36. <u>(Amended)</u> The analytical apparatus of claim 35 wherein the ~~syringe comprises a~~ plunger driven by the control and management system through a ~~precision~~ translation mechanism, enabling precision volume control.

37. (Amended) The analytical apparatus of claim 36 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor.

38. (Amended) The analytical apparatus of claim 27 wherein individual ones of the syringes in the system of syringes are connected by switching valves to individual inputs to a mixer having a single output, such that, with one syringe holding a precise volume of an isotope mixture at a first concentration and a second syringe holding a precise volume of the solvent, driving the plungers of the two syringes simultaneously creates an isotope mixture at the single output of a ~~precise expected~~predetermined concentration less than the concentration of the mixture in the one syringe.

40. (Amended) The extraction module of claim 39 wherein the ~~raw-sample~~raw sample reservoir is generally vertically-oriented and the conduit from the liquid systems enters at a height on the reservoir such that a liquid sample drawn into the reservoir may have a gas gap over the sample.

43. (Amended) The extraction module of claim 39 wherein the syringe comprises a plunger with a ~~precision~~ translation mechanism drivable by a control and management system, enabling precision volume control.

44. (Amended) The extraction module of claim 43 wherein the ~~precision~~ translation mechanism is controllable at variable translation rates, enabling variation of fluid transfer rate.

45. (Amended) The extraction module of claim 43 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor.

46. (Amended) The extraction module of claim 39 further comprising a flush reservoir connected through additional valves to the syringe, to the ~~raw-sample~~raw sample reservoir, to a source of flushing liquids, to a second vacuum source, to a cover-gas source, and to a drain line, such that the elements of the extraction module may be flushed and cleaned between sample cycles.

47. <u>(Amended)</u> In an analytical apparatus to monitor liquid systems, the apparatus having modules for extracting samples, modifying the samples, and analyzing the samples for concentration of specific constituents, a chemical ~~alteration~~<u>modification</u> module comprising:

a chemical reservoir having a solution of precisely compounded chemicals; and

apparatus for providing portions of the solution to other elements of the analytical apparatus.

49. <u>(Amended)</u> The chemical alteration module of claim 48 wherein the syringe comprises a plunger having a ~~precision~~ translation mechanism drivable by a control and management system, enabling precision volume control.

50. <u>(Amended)</u> The chemical alteration module of claim 49 wherein rate of translation of the ~~precision~~ translation mechanism is variable, enabling variation of rate of liquid transfer.

51. <u>(Amended)</u> The chemical alteration module of claim 49 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor.

54. <u>(Amended)</u> The isotope dilution module of claim 53 wherein the mechanism for providing portions of the ~~solution~~<u>mixture</u> comprises a syringe connected by intermediate conduit to the at least one isotope reservoir below a surface of the mixture in the reservoir, such that withdrawing a plunger of the syringe by a precise distance draws a precise volume of the mixture in the isotope reservoir into the syringe.

55. <u>(Amended)</u> The isotope dilution module of claim 54 wherein the syringe comprises a plunger having a ~~precision~~ translation mechanism drivable by a control and management system, enabling precision volume control.

56. <u>(Amended)</u> The isotope dilution module of claim 55 wherein the control and management system is ~~enabled~~<u>structured</u> to vary <u>the</u> rate of plunger translation.

57. <u>(Amended)</u> The isotope dilution module of claim 55 wherein the ~~precision~~ translation

mechanism comprises a ~~precision~~ stepper motor.

59. (Amended) The isotope dilution module of claim 54 wherein the isotope reservoir contains an isotope mixture in a solvent at a specific concentration, and further comprising a solvent reservoir in addition to the at least one isotope reservoir, the solvent reservoir containing a solvent common to the isotope mixture in the isotope reservoir, and a system of syringes, connecting conduits and mixers, enabling portions of the isotope mixture to be diluted ~~to lesser concentrations~~ before being provided to other elements of the analytical apparatus.

60. (Amended) The isotope dilution module of claim 59 wherein the isotope reservoir holds an isotope mixture in a concentration ~~if~~of about 1 part ~~isotope~~isotopes in one million parts of the mixture ~~(1ppm)~~, and the system of syringes, connecting conduits and mixtures enables dilution of six orders of magnitude to about one part per trillion ~~(1ppt)~~.

63. (Amended) The isotope dilution module of claim 62 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor.

64. (Amended) The isotope dilution module of claim 59 wherein individual ones of the syringes in the system of syringes are connected by switching valves to individual inputs to a mixer having a single output, such that, with one syringe holding a precise volume of an isotope mixture at a first concentration and a second syringe holding a precise volume of the solvent, driving the plungers of the two syringes simultaneously creates an isotope mixture at the single output of a ~~precise expected~~predetermined concentration less than the concentration of the mixture in the one syringe.

65. (Amended) A method for monitoring fluid systems for concentration of selected species, comprising the steps of:

(a) drawing raw samples of the fluid systems one-at-a-time into at least one ~~raw-sample~~raw sample reservoir connected by input fluid conduit to individual ones of the fluid systems;

(b) modifying the raw samples by addition of material from one or more modification modules;

(c) providing measured portions of modified samples to an analytical device; and

(d) determining concentration of the selected species by a controller that receives output from the analytical device.

68. (Amended) The method of claim 67 wherein the wet baths are baths in a semiconductor fabrication unit ~~(fab)~~ devoted to semiconductor integrated circuit ~~(IC)~~ manufacturing.

75. (Amended) The method of claim ~~65~~61 wherein, in step (a), a gas gap is maintained over a liquid sample drawn into the sample reservoir, by implementing the ~~raw sample~~raw sample reservoir as a generally vertically-oriented container with conduit from the wet-bath entering at a height on the reservoir above a level anticipated for a sample volume in the reservoir.

79. (Amended) The method of claim 78 wherein the syringe is operated by a plunger driven through a ~~precision~~ translation mechanism, enabling ~~precision~~ volume control.

81. (Amended) The method of claim 80 wherein the ~~precision~~ translation mechanism comprises a ~~precision~~ stepper motor, and rate of translation is controlled by controlling the stepping rate of the motor.~~.~~

84. (Amended) The method of claim 65 wherein, in step (b), chemicals are added in solution to samples from chemical modification~~alteration~~ modules comprising at least one chemical reservoir having a solution of precisely compounded chemicals and apparatus for providing portions of the solution to the samples.

86. (Amended) The method of claim 85 wherein the syringe is operated by a plunger driven through a ~~precision~~ translation mechanism, enabling precision volume control.

88. (Amended) The method of claim 86 wherein the ~~precision~~ translation mechanism is driven by a ~~precision~~ stepper motor.

90. (Amended) The method of claim ~~89~~92 wherein ~~the~~ portions of the sample~~provided to~~

~~other elements~~ are provided through a mixer having two or more inputs to one output, with one input connected to the chemical ~~alteration~~<u>modification</u> module and another connected to another module in the analytical apparatus.

94. <u>(Amended)</u> The method of claim 93 wherein the syringe is operated by a plunger driven through a ~~precision~~ translation mechanism, enabling precision volume control.

96. <u>(Amended)</u> The method of claim 95 wherein the rate is varied by controlling the step rate of a ~~precision~~ stepper motor driving the translation mechanism.

100. <u>(Amended)</u> The method of claim 92 wherein isotope mixtures are provided in different concentration levels by maintaining an isotope mixture in a solvent at a specific concentration in the isotope reservoir, and diluting portions of that mixture with solvent from a solvent reservoir, the solvent reservoir containing the solvent common to the isotope mixture in the isotope reservoir, doing the dilution through a system of syringes, connecting conduits and mixers, enabling portions of the isotope mixture to be diluted ~~to lesser concentrations~~ before being provided to other elements of the analytical apparatus.

101. <u>(Amended)</u> The method of claim 100 wherein the isotope reservoir holds an isotope mixture in a concentration ~~if~~<u>of about</u> 1 part ~~isotope~~<u>isotopes</u> in one million parts of the mixture ~~(1ppm)~~, and the system of syringes, connecting conduits and mixtures enables dilution of six orders of magnitude to <u>about</u> one part per trillion ~~(1ppt)~~.

103. <u>(Amended)</u> The method of claim 102 wherein the plunger of an individual syringe is driven through a ~~precision~~ translation mechanism, enabling precision volume control.

104. <u>(Amended)</u> The method of claim 103 wherein the precision ~~translation~~ mechanism is driven by a precision stepper motor.

105. <u>(Amended)</u> The method of claim 103 wherein individual ones of the syringes in the system of syringes are connected by switching valves to individual inputs to a mixer having a single output, such that, with one syringe holding a precise volume of an isotope mixture at a first concentration and a second syringe holding a precise volume of the solvent,

driving the plungers of the two syringes simultaneously creates an isotope mixture at the single output of a precise ~~expected~~predetermined concentration less than the concentration of the mixture in the one syringe.